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## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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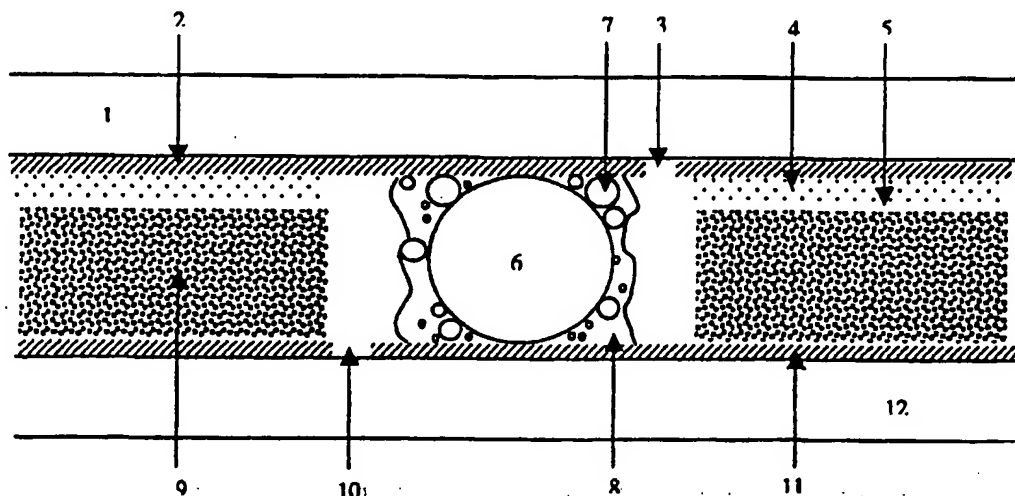
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With international search report.  
With amended claims.

(54) Title: METHODS TO IMPLEMENT INTERCONNECTS IN MULTI-CELL REGENERATIVE PHOTOVOLTAIC PHOTOELECTROCHEMICAL DEVICES



## (57) Abstract

The present invention relates to multi-cell regenerative photovoltaic photoelectrochemical (RPEC) devices. The invention describes the structure of a multi-cell RPEC device where conductive interconnects are formed by a matrix mounting conductive particles, formed between extended portions of opposed planar conductive members.

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METHODS TO IMPLEMENT INTERCONNECTS IN MULTI-CELL  
REGENERATIVE PHOTOVOLTAIC PHOTOELECTROCHEMICAL DEVICES

TECHNICAL FIELD

5           This invention relates to multi-cell regenerative  
photovoltaic photoelectrochemical (RPEC) devices,  
materials and methods used for internal electrically  
conductive connections (here called 'interconnects') for  
such devices, and materials and methods used for dividing  
10 electrically conducting layers within such devices.

Examples of the RPEC cells of the type concerned are  
disclosed in the following US patents:

4927721, Photoelectrochemical cell; Michael Graetzel  
and Paul Liska, 1990.

15           5350644, Photovoltaic cells; Michael Graetzel,  
Mohammad K Nazeeruddin and Brian O'Regan, 1994.

5525440, Method of manufacture of photo-  
electrochemical cell and a cell made by this method;  
Andreas Kay, Michael Graetzel and Brian O'Regan, 1996.

20           5728487, Photoelectrochemical cell and electrolyte  
for this cell; Michael Graetzel, Yordan Athanasov and  
Pierre Bonhote, 1998.

BACKGROUND TO THE INVENTION

25           Photoelectrochemical PV cells, as of the type  
disclosed in the above patents, are capable of being  
fabricated in a laminate arrangement between two large  
area substrates without undue expense. A typical  
arrangement utilises electrically conducting coatings upon  
30 the internal surfaces of such substrates, with at least  
one of such substrates being transparent to visible light  
(eg. comprised of glass or plastics) and coated with a  
transparent electron conductor (TEC). However, such TEC

coatings, which usually comprise a metal oxide(s), have high resistivity when compared with normal metal conductors, resulting in high resistive losses for large area RPEC cells. In addition, individual RPEC cells  
5 generate a voltage that is inadequate for many applications. A multiple of RPEC cells (here called 'RPEC modules') connected in series would generate higher voltages and minimise total current, thereby minimising power loss due to the resistance of such TEC coating(s).  
10 External series connection of RPEC cells can increase manufacturing costs and introduce additional resistive losses. To enable internal series connection of adjacent RPEC cells, selected areas of such conducting coatings must be electrically isolated, portions of such areas  
15 overlapped when laminated, interconnects used to connect such overlapped areas and electrolyte-impermeable barriers used to separate the electrolyte of individual cells.

#### SUMMARY OF THE INVENTION

20 Preferably, the present invention provides materials and methods for interconnects for use with RPEC modules that will overcome the mentioned disadvantages in the art.

The present invention provides a regenerative photovoltaic  
25 photoelectrochemical device comprising two or more photoelectrochemical cells laminated between two substrates (1,12), with opposing electrical conductors (2,11) supported upon the internal surfaces of said substrates, wherein at least one conductor and it's  
30 adjacent substrate is substantially transparent to visible light, and wherein said conductors on each substrate are each divided into electrically isolated regions with each said cell being formed between parts of two regions of

said opposing planar conductors and with each said cell comprising: a photoanode (3) a cathode (11) and an electrolyte medium (5), located between said photoanode and said cathode, where said adjacent photoelectrochemical  
5 cells are electrically interconnected in series by an electrically interconnecting material comprising conductive particles (6).

Note that interconnects for use with RPEC modules preferably need not have conductivities approaching that  
10 of metals, because the interconnection is made along the entire length of each cell and, moreover, the conduction path length is normally only 30 $\mu$ m-50 $\mu$ m, which is the distance between opposing electrically conducting coatings. Thus we have found that efficient and  
15 satisfactory RPEC modules can be made with interconnects having bulk resistivity below 20 ohm cm.

In one embodiment, this invention involves the use of an improved composite material as such interconnects which can be deposited as a thin strip of liquid or paste, so  
20 that such a strip is adapted to bridge such overlapped areas of conducting coatings and then cured (e.g. cross-linked), thermoset, dried, sintered or otherwise processed to form an electronic conductor between such conducting coatings after such opposing substrates have been  
25 assembled. Whilst said composite material can be deposited by conventional screen or stencil printing, this process may cause damage to electrodes previously deposited. In one preferred embodiment of the invention, the composite material is deposited as a thin strip of  
30 liquid or paste from a nozzle, whereby said nozzle or substrate or both are moved to effect such deposition. The composite material preferably comprises electrically conducting particles embedded in a polymeric matrix. In a

preferred embodiment of the invention, the relative proportion by volume of conductive particles to matrix material in said interconnects is preferably between 1:5 and 2:1, with the high concentrations of conductive  
5 particles being preferred.

In one preferred embodiment of the invention, said interconnect may be substantially impervious to and unreactive towards the electrolyte of the RPEC cells, thereby also performing the function of an electrolyte-  
10 impermeable barrier. In another preferred embodiment of the invention, the interconnect may be unreactive towards, but not substantially impervious to, the electrolyte of the RPEC cells. In this preferred embodiment of the invention, an electrolyte-impermeable barrier located  
15 beside the interconnect is used to separate the electrolyte of individual cells. In this preferred embodiment of the invention, said conductive particles and said polymer matrix of said interconnect may be selected from a wider range of materials due to less stringent  
20 chemical permeability requirements. In another preferred embodiment of the invention, the interconnect is chemically isolated from the electrolyte of the RPEC cells by electrolyte-impermeable barriers on both sides of the interconnect. In this preferred embodiment of the  
25 invention, said conductive particles and said polymer matrix of said interconnect may be selected from a wider range of materials due to less stringent chemical reactivity requirements. Said impermeable barriers may be electrically conducting or non-conducting and may be  
30 composed of any suitable material, including, but not limited to, silicones, epoxies, polyesters, polyolefins, acrylic, ormocers and thermoplastics. Said impermeable barriers may be deposited as thin strips of liquid or

paste. It is preferable that said impermeable barriers are co-deposited with said interconnect from separate nozzles mounted beside the nozzle from which said interconnect is deposited. In another preferred embodiment of the invention, the interconnect comprises said composite material with a very thin strip of conductive polymer located between said composite material and one or both said conducting coatings. In this preferred embodiment of the invention, said very thin strip(s) of conductive polymer provides improved electrical conductivity between said composite material and said conducting coating(s). In this preferred embodiment of the invention, said very thin strip(s) of conductive polymer may contain polypyrroles, polyanalines, 3,4-ethylene dioxythiophenes and the like and is preferably deposited from a nozzle as previously described.

The polymeric matrix of the interconnect may be electrically conducting (e.g. containing polypyrroles, polyanalines, 3,4-ethylene dioxythiophenes and the like) or may be electrically insulating (e.g. containing silicones, epoxies, polyesters, polyolefins, acrylics, ormocers, thermoplastics). Suitable materials for conducting particles may include but are not limited to metallic conductors such as metallic materials (e.g. tungsten, titanium and platinum) in the form of particles and/or metallic beads, and non-metallic conductors such as carbon, ceramics (e.g. indium tin oxide, ruthenium dioxide, cadmium stannate and fluorine-doped stannic oxide), and conducting polymers (e.g. polypyrroles, polyanalines, 3,4-ethylene dioxythiophenes and the like, which can be formed as particles from solution or formed in bulk and commuted to size).



In one preferred embodiment of the invention, the conducting particles are of dimensions approximating the distance between the opposing electrically conducting coatings. In another preferred embodiment of the invention, the conducting particles are of dimensions smaller than said distance and are preferably of dimensions less than 5% of said distance. In still another preferred embodiment of the invention, the conducting particles are of a mixture of sizes, whereby some said particles are of dimensions approximating said distance and other said particles are of dimensions smaller than said distance and are preferably of dimensions less than 5% of said distance. In still another preferred embodiment of the invention, the conducting particles are of a mixture of sizes, whereby some said particles, comprising at least 20% of the total weight of such particles, are of dimensions approximating said distance and other said particles are of dimensions smaller than said distance and are preferably of dimensions less than 5% of said distance. In still another preferred embodiment of the invention, the conducting particles are of a mixture of sizes, whereby some said particles are of dimensions approximating said distance and other said particles, comprising at least 20% of the total weight of such particles, are of dimensions smaller than said distance and are preferably of dimensions less than 5% of said distance. In still another preferred embodiment of the invention, the conducting particles are of a mixture of sizes, whereby some said particles are of dimensions approximating said distance, other said particles are of dimensions smaller than said distance and are preferably of dimensions less than 5% of said distance, and still other said particles

are of dimensions less than said distance and greater than 5% of said distance. Certain mechanical forces, such as wind loading, may cause a short circuit to occur between the laminated electrodes in some RPEC module designs. It is a feature of this invention, that interconnects containing conductive particles of dimensions approximating to or near to said distance between said opposing electrically conducting coatings may prevent or minimise the occurrence of such short circuits during such mechanical loading.

#### BRIEF DESCRIPTION OF DRAWINGS

Having broadly portrayed the nature of the present invention, embodiments thereof will now be described by way of example and illustration only. In the following description, reference will be made to the accompanying drawings in which:

Figure 1 is an enlarged partial transverse section of the region surrounding an interconnect within a RPEC module formed in accordance one example of the invention.

Figure 2 is a diagrammatic perspective representation of a RPEC module formed in accordance with the above example of the invention.

Figure 3 is an enlarged partial transverse section of the region surrounding an interconnect within an RPEC module formed in accordance with a further example of the invention.

Referring to Figure 1, this portion of the RPEC module comprises two glass substrates (1,12), both of which are coated with a transparent electron conductor (TEC) coating (2,11). The TEC layers are selectively isolated (3,10) in order to electrically separate each individual cell. The cathode comprises a platinum

electrocatalyst (4) attached to one TEC coating (2), the photoanode comprises ruthenium dye-sensitised titania (9) attached to the other TEC coating (11), and an electrolyte (5) containing a redox mediator is located between the cathode and photoanode. All of the aforementioned materials are described in more detail in US Patent 5350644. The interconnect is comprised of two different electrically conducting particles, 45um titanium (6) and 0.5um tungsten (7), embedded within a polymeric silicone matrix (8), in proportions by weight of 1 to 10 to 4 respectively.

Referring to Figure 2, this diagram of a RPEC module comprises two glass substrates (1,12), both of which are coated with a transparent electron conductor (TEC) coating (2,11). The cathode comprises a platinum electrocatalyst (4) attached to one TEC coating (2). The photoanode comprises ruthenium dye-sensitised titania (9) attached to the other TEC coating (11). Also shown are the interconnect (13) and the strips where the TEC coating has been removed (3,10).

Referring to Figure 3, a further example of a RPEC module formed in accordance with the present invention is illustrated. The same reference numerals have been used in figure 3 for the same components as designated in figure 1 and no further description will be given of these components. Reference numeral (14) designates protective layers forming electrolyte-impermeable barriers for separating the electrolyte (5) of individual cells.

THE CLAIMS DEFINING THE INVENTION ARE AS FOLLOWS:What is claimed:

- 5 1. A regenerative photovoltaic photoelectrochemical device comprising two or more photoelectrochemical cells laminated between two substrates (1,12), with  
opposing electrical conductors (2,11) supported upon  
the internal surfaces of said substrates, wherein at  
10 least one conductor and it's adjacent substrate is  
substantially transparent to visible light, and  
wherein said conductors on each substrate are each  
divided into electrically isolated regions, with each  
said cell being formed between parts of two regions  
15 of said opposing planar conductors and with each said  
cell comprising: a photoanode (3) a cathode (11) and  
an electrolyte medium (5), located between said  
photoanode and said cathode, where said adjacent  
photoelectrochemical cells are electrically  
20 interconnected in series by an electrically  
interconnecting material comprising conductive  
particles (6).
- 25 2. The photoelectrochemical device in accordance with  
claim 1, wherein the conductive particles are placed  
between a separate part of the region and of said  
conductor adjacent said photoanode of n-th said cell  
and a separate part of the region of said opposing  
conductor adjacent to said cathode of n-th +1th said  
30 cell.
3. The photoelectrochemical device according to claim 1  
or claim 2, wherein external electrical connections

are made to said device via electrical contacts (7) placed upon a separate part of the region of said conductor adjacent to said cathode of the first said cell and upon a separate part of the region of said conductor adjacent to said photoanode of the last said cell.

4. The photoelectrochemical device of claim 2 or claim 3, wherein said electrically conductive particles include particles of dimensions approximating the distance between said opposing conductors.
5. The photoelectrochemical device of any one of the preceding claims, wherein said electrically interconnecting material is comprised of the conductive particles contained within a matrix.
6. The photoelectrochemical device of claim 5, wherein the matrix is a polymeric matrix.
7. The photoelectrochemical device of any one of claims 2 to 6, wherein said electrically conductive particles include particles of dimensions smaller than the distance between said opposing conductors.
8. The photoelectrochemical device of claim 7, wherein the electrically conductive particles are of dimensions less than 5% of said distance between said opposing conductors.

9. The photoelectrochemical device of any one of claim 2 to 6, wherein said electrically conductive particles are of a mixture of sizes, whereby some of said electrically conductive particles are of dimensions approximating the distance between said opposing planar conductors and other said electrically conductive particles are of dimensions smaller than said distance.
10. 10. The photoelectrochemical device of claim 9, wherein the dimension of the other said electrically conductive particles are less than 5% of the distance between said opposing conductors.
11. 11. The photoelectrochemical device of any one of claims 6 to 10 wherein said polymeric matrix is wholly or partially comprised of one or more electrically conducting polymer materials.
12. 12. The photoelectrochemical device according to any one of the preceding claims wherein the conductive particles are made of Pt, W, Ti or carbon.
13. 13. The photoelectrochemical device according to any one of the preceding claims wherein the conductive particles are made of conductive oxides such as ITO, Ru - oxide;
14. 14. The photoelectrochemical device of any one of the preceding claims, wherein an additional layer of conductive polymer is placed between said conductive particles and the conductors

15. The Photoelectrochemical device of anyone of the preceding claims, wherein the photoanode consists of a layer of large band gap semiconductor (3), which is sensitised by an extremely thin, adsorbed layer of dye (4) that absorbs a large fraction of visible light.
16. The photoelectrochemical device of anyone of the preceding claims, an electro catalytically active material (10) being deposited on the cathode (11)
17. The photoelectrochemical device of anyone of the preceding claims, the electrolyte medium (5) containing redox mediator.

**AMENDED CLAIMS**

[received by the International Bureau on 07 July 2000 (07.07.00) ;  
original claims 1-17 replaced by new claims 1-16 (4 pages)]

**THE CLAIMS DEFINING THE INVENTION ARE AS FOLLOWS:****What is claimed:**

- 5 1. A regenerative photovoltaic photoelectrochemical device  
comprising two or more photoelectrochemical cells  
laminated between two substrates (1,12), with opposing  
electrical conductors (2,11) supported upon the  
internal surfaces of said substrates, wherein at least  
10 one conductor and it's adjacent substrate is  
substantially transparent to visible light, and wherein  
said conductors on each substrate are each divided into  
electrically isolated regions, with each said cell  
being formed between parts of two regions of said  
15 opposing planar conductors and with each said cell  
comprising: a photoanode (3) a cathode (11) and an  
electrolyte medium (5), located between said photoanode  
and said cathode, and said adjacent  
photoelectrochemical cells are electrically  
20 interconnected in series by an electrically  
interconnecting material comprising conductive  
particles (6), where said electrically conductive  
particles include particles of dimensions approximating  
the distance between said opposing conductors.
- 25 2. The photoelectrochemical device in accordance with  
claim 1, wherein the conductive particles are placed  
between a separate part of the region and of said  
conductor adjacent said photoanode of n-th said cell  
30 and a separate part of the region of said opposing  
conductor adjacent to said cathode of n-th +1th said  
cell.



3. The photoelectrochemical device according to claim 1  
or claim 2, wherein external electrical connections are  
made to said device via electrical contacts (7) placed  
upon a separate part of the region of said conductor  
adjacent to said cathode of the first said cell and  
upon a separate part of the region of said conductor  
adjacent to said photoanode of the last said cell.
4. The photoelectrochemical device of any one of the  
preceding claims, wherein said electrically  
interconnecting material is comprised of the conductive  
particles contained within a matrix.
5. The photoelectrochemical device of claim 4, wherein the  
matrix is a polymeric matrix.
6. The photoelectrochemical device of any one of claims 2  
to 5, wherein said electrically conductive particles  
include particles of dimensions smaller than the  
distance between said opposing conductors.
7. The photoelectrochemical device of claim 6, wherein the  
electrically conductive particles are of dimensions  
less than 5% of said distance between said opposing  
conductors.
8. The photoelectrochemical device of any one of claim 2  
to 5, wherein said electrically conductive particles  
are of a mixture of sizes, whereby some of said  
electrically conductive particles are of dimensions  
approximating the distance between said opposing planar  
conductors and other said electrically conductive  
particles are of dimensions smaller than said distance.

9. The photoelectrochemical device of claim 8, wherein the dimension of the other said electrically conductive particles are less than 5% of the distance between said opposing conductors.
10. The photoelectrochemical device of any one of claims 5 to 9 wherein said polymeric matrix is wholly or partially comprised of one or more electrically conducting polymer materials.
11. The photoelectrochemical device according to any one of the preceding claims wherein the conductive particles are made of Pt, W, Ti or carbon.
12. The photoelectrochemical device according to any one of the preceding claims wherein the conductive particles are made of conductive oxides such as ITO, Ru - oxide;
13. The photoelectrochemical device of any one of the preceding claims, wherein an additional layer of conductive polymer is placed between said conductive particles and the conductors
14. The photoelectrochemical device of anyone of the preceding claims, wherein the photoanode consists of a layer of large band gap semiconductor (3), which is sensitised by an extremely thin, adsorbed layer of dye (4) that absorbs a large fraction of visible light.

15. The photoelectrochemical device of anyone of the preceding claims, an electro catalytically active material (10) being deposited on the cathode (11)

5

16. The photoelectrochemical device of anyone of the preceding claims, the electrolyte medium (5) containing redox mediator.

10

Figure 1

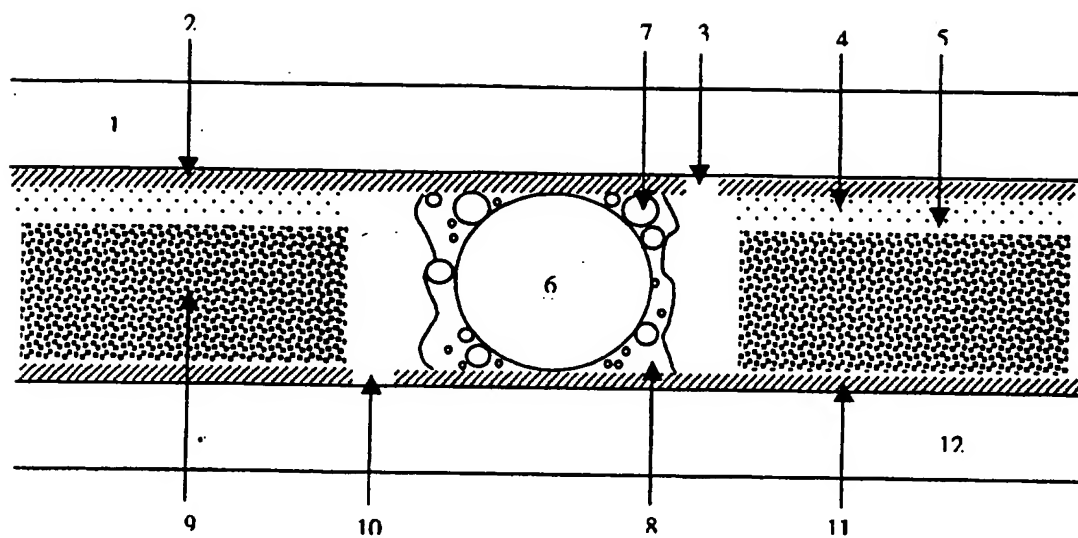


Figure 2

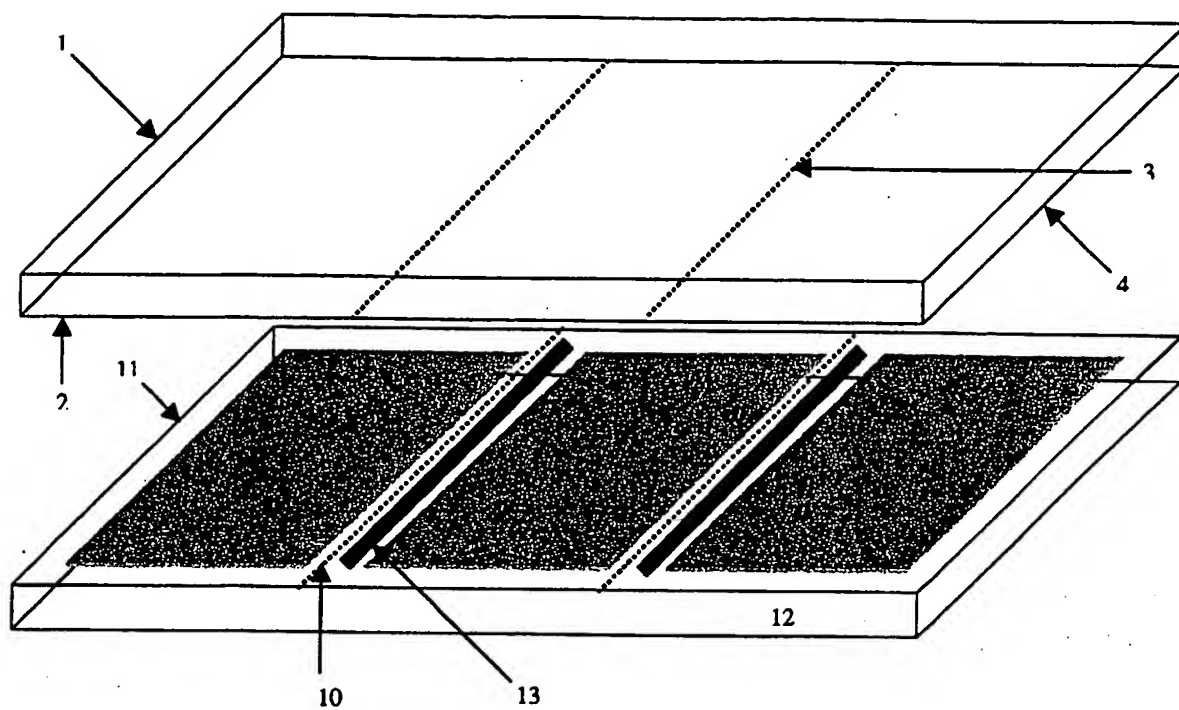
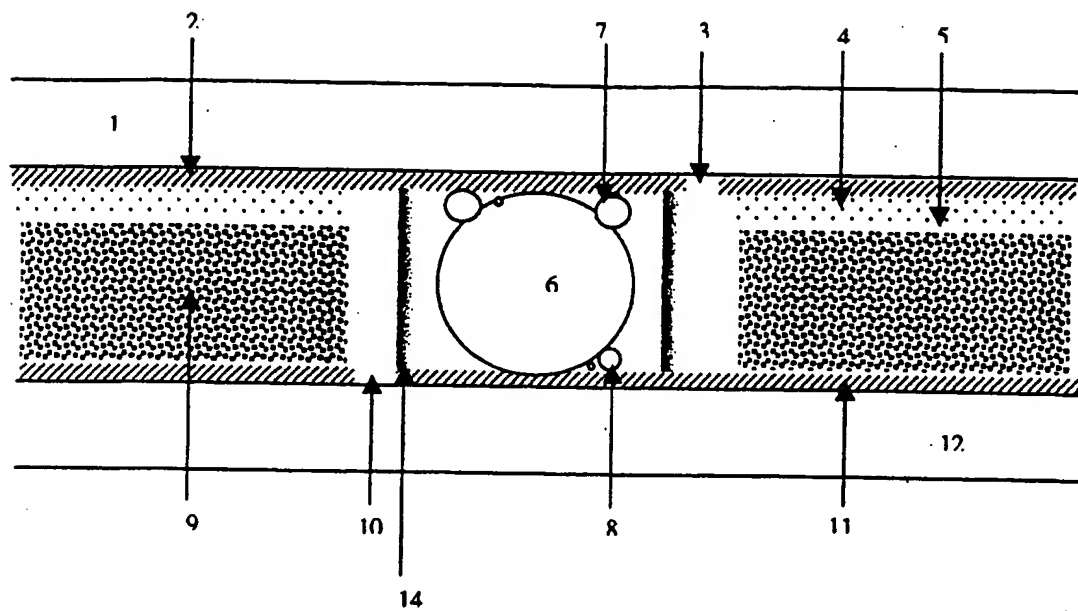


Figure 3



## INTERNATIONAL SEARCH REPORT

International application No.

PCT/AU00/00190

**A. CLASSIFICATION OF SUBJECT MATTER**Int. Cl. <sup>7</sup>: H01G 9/20, 9/004, 9/26, H01M 6/36

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)

H01G 9/-, H01M, H01L 31/-

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)  
DWPI, JAPIO with keywords (RPEC or photoelectro+ or photo electro+) and conduct+ and (particle? or grain?)**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	DE 4306404 A1(ABB Research Ltd) 8 September 1994 See the abstract, col 2 line 10 to col 3 line 20	1-17
X	DE 4416247 A1 (KAY) 9 November 1995 See the abstract, col 2 line 35 to col 3 line 28	1-17
X	DE 19540712 A1 (Ecole Polytechnique Federale de Lausanne) 7 May 1997 See the abstract, col 3 line 57 to col 4 line 3	1-17

☒ Further documents are listed in the continuation of Box C
 ☐ See patent family annex

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4 May 2000

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10 MAY 2000

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## INTERNATIONAL SEARCH REPORT

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C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	DE 4412050 A1 (Hoffmann et al) 5 January 1995 See the abstract	
A	DE 19528401 A1 (Fraunhofer-Gesellschaft zur Forderung der angewandten Forschung e.V.) 6 February 1997 See the abstract	